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L SUBSHELL RATIOS OF E2 TRANSITIONS IN DEFORMED RARE EARTH NUCLEI*

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ABSTRACT The L subshell ratios of the $2+$ to $0+$ first excited state to ground state transitions in Gd^{154}, Yb^{170} and W^{182} have been measured with an iron-free double-focusing spectrometer. The results indicate that the L_1/L_2 and L_1/L_3 ratios are about 15% or more higher than the theoretical values while L_2/L_3 ratios agree with the theories of Rose, and of Sliv and Band

INTRODUCTION

A powerful tool for the determination of gamma transition-multipolarities is the comparison of experimental and theoretical L-subshell intensity ratios. The accuracy of the multipolarities and mixing ratios obtained in this manner depends on the validity of the theoretical conversion-coefficient ratios used in the analysis On a visit to Vanderbilt University in May, 1964, M Mladjenovic communicated some results of studies being carried out in Belgrade on the measurements of L-subshell ratios of $2+$ to $0+$ pure E2 transitions in the deformed rare-earth region These results did not agree with the theoretical ratios either of Rose (1958) or of Sliv and Band (1956) and Sliv (1961). The L_1/L_2 and L_1/L_3 ratios were found to be ten to thirty percent higher than theory while L_2/L_3 agreed. Following his communication, the present work was begun on the measurements of L-subshell ratios in the rare-earth region Later, at the International Conference on the Internal Conversion Process held at Vanderbilt University, May 10-13, 1965, other groups Stepic *et al.*, Novakov *et al.*, and Karlsson *et al.*, presented experimental data on L-subshell ratios which also disagreed with theoretical results. A brief account of the results of the work described in the present paper were presented at the conference, Brantley *et al.*, (1965). The decay schemes of Eu^{154}, Tm^{170} and Ta^{182} are shown in figures 1, 2, and 3. The transitions studied were the first excited state to ground state $2+$ to $0+$, transitions in each case.

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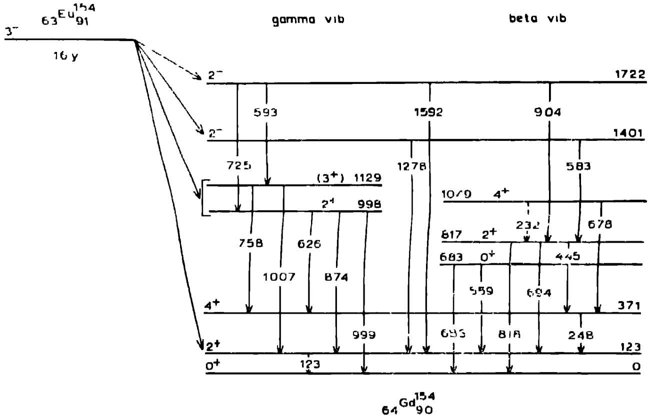

Fig. 1. Decay scheme of Eu^{154} from Reidinger et al (1965) and Brantley et al (1966)

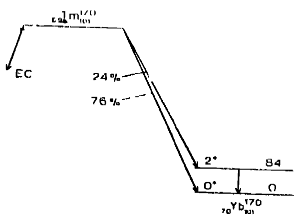

Fig. 2 Decay scheme of Tm^{170} as given in Nuclear Data Sheets

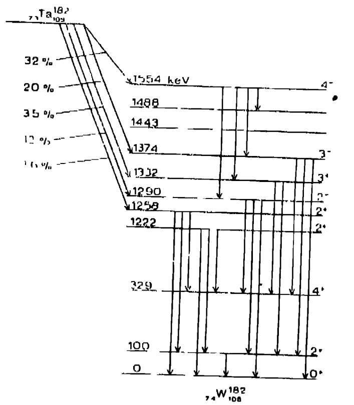

Fig 3 Decay scheme of Ta^{182} with only the important feature as given in the Nuclear Data Sheets and recent literature see for example Daniel et al

EXPERIMENTAL TECHNIQUES AND SOURCE PREPARATION

These measurements were done on a 30-cm iron-free, double-focusing spectrometer, Baird *et al.*, (1962). Momentum resolutions better than about 0.10 percent were required for the present studies The best previous resolution obtained on the spectrometer was 0.13 percent, which was thought to be near the limit obtainable on the spectrometer However, the resolution was improved by the use of narrower sources and by better alignment of the source in the spectrometer. A narrow defining slit was placed over the sources The resolution for the various measurements varied from about 0.06 percent to 0.10 percent This resolution is near the limit of the control mechanism of the spectrometer and care must be taken to insure that no instrumental distortions enter into the measurements Several different measurements for each transition were done and care was taken to assure optimum current control in the instrument over the long counting periods of 24-48 hours A continuous gas-flow G M. counter with a 0.37-mg/cm² aluminum-coated Mylar window was used. The cutoff energy of the window, about 11 keV, was sufficiently low to insure 100 percent transmission at the energies at which the measurements were taken The lowest energy of the three transitions measured is 84.2 keV for which the L_1 line is found at about 73 keV.

The Eu^{154} activity was obtained by irradition of high-purity Eu^{153} in the Materials Testing Reactor at Idaho Falls. The activity was then allowed to decay for nine months so that the short-lived components died out or became negligible In particular, Eu^{156} produced by triple neutron capture in the high flux of the reactor (about 5.6×10^{14} neutrons/cm²-sec) has a half life of only fifteen days, so that the amount of it remaining in the sample is negligible From a consideration of its decay scheme one concludes that the 1.81*y* Eu^{155} can also be neglected Twenty-five percent of the decay of Eu^{155} is to the ground and first excited state of Gd^{155}, and since the energy of the first excited state of Gd^{155} is only 60 keV this part of the decay can produce no transitions which would interfere with the work on Gd^{154}. Almost all of the remainder of the Eu^{155} decay is to the second and third excited states of Gd^{155} at 87 and 105 keV, so again no transitions of sufficient energy to interfere with the Gd^{154} work occur from these decays. Internal-conversion spectra and gamma-ray spectra taken with a solid-state detector by Riedinger *et al* (1965) were used to determine the 13*y* Eu^{152} contamination The Eu^{152} contamination was estimated to be less than one percent The Eu^{154} specific activity was 34.4 milli-curies/milli-gram The source was prepared by thermal evaporation from the oxide on to a one-mil platinum backing The dimensions of this source, as well as those of the next two sources discussed, were determined by an aluminum defining slit placed close to the sources. The surce had dimensions of 0.4×18 mm² The source thickness was estimated to be 2 micro-grams/centimeter from knowledge of the specific activity and the amount used and from the intensity of the L_2 line of the 123 keV transition combined with knowledge of the conversion coefficients and the spectrometer transmission

Difficulty was encountered in fabrication of the Ta^{182} source because of the high melting point of tantalum and the difficulties of electroplating this material. The Ta^{182} activity was obtained from Oak Ridge National Laboratory in the chemical form of tantalate in a KOH solution. Its specific activity was 3.0 milli-curies/milli-gram. Thermal evaporation of the Ta^{182} in vacuum was impossible because the boiling point of the tantalum is above the melting point of the crucible. This problem was solved by deposition of the activity in solution (tantalate in KOH) onto a thin strip of platinum. The platinum strip was then placed in vacuum and a current was passed through the strip, so the excess mass of the dried solution evaporated and left behind the Ta^{182} activity. The source dimensions were 0.4×18 mm^2 and its thickness less than 15 micro-grams/centimeter^2 obtained with the same procedure as for the Eu^{154}.

The Tm^{170} activity was also obtained from Oak Ridge National Laboratory. Its chemical form was $TmCl_3$ in weak HCl. Its specific activity was 80.7 milli-curies/milli-grams. The Tm^{170} activity was liquid-deposited onto one-tenth mil Mylar which was coated with aluminium on one side. The source thickness was estimated in the same manner as described for Eu^{154} to be less than 10 micro-grams/centimeter^2.

The requirement of good resolution made it necessary that the sources have minimum mass and be 1 mm wide or less. To achieve this narrow width, an aluminium disk with a narrow defining slit was placed over the sources. The dimensions of this defining slit were 0.4×18 mm^2. The counter window was of the same width. This defining slit was found to improve the resolution without distorting the spectrum. Measurements were made with and without the slit and there was no apparent effect on the data other than the improvement in resolution.

RESULTS AND DISCUSSION

The L subshell lines of the 123-keV transition in Gd^{154} were measured five times and the results averaged. The resolution for the different runs was varied from 0.06 to 0.10 percent by variation of the spectrometer baffles. In order to separate the L_1 line from the L_2 line in the analysis, the line shape of the L_3 line was used as a standard. It was normalized to the L_2 line, and the L_1 line was subtracted. A typical measurement is shown in figure 5. The results are presented in Table I. The L_1/L_2 and L_1/L_3 ratios are higher than theory, while the L_2/L_3 ratio agrees with theory. Because the L_1/L_2 and L_1/L_3 ratios are comparable, a convenient method of showing the results is to average the experimental value of these two quantities and divide this value by the average of the same quantities for the two theories. The mean percentage deviation is obtained by subtracting one from this quotient and multiplying the result by 100. The mean percentage deviation for the Gd^{154} case is 17 ± 7 higher than theory.

The L subshell lines of the 84 keV transition in Yb^{170} were measured two times and the results averaged. The resolution on one run was about 0.06 percent and on the other run about 0.08 percent. In these measurements line tailing due to source thickness was a problem. In order to minimize the errors which are in-

TABLE I

L Subshell ratios for $2+ \rightarrow 0+$ Pure E2 transitions

Nuclide	E_γ keV	Exptl. Ratios $\times 10^3$ L_1/L_2	L_1/L_3	L_2/L_3	Rose $\times 10^3$ L_1/L_2	L_1/L_3	L_2/L_3	Sliv and Band $\times 10^3$ L_1/L_2	L_1/L_3	L_2/L_3	$\frac{(L_2/L_3)\text{ expt}}{(L_2/L_3)\text{th av}}$	$\frac{(L_1/L_2+L_1/L_3)\text{ expt}}{(L_1/L_2-L_1/L_3)\text{th av.}}$	Reference
$_{64}Gd^{154}$	123.1	397±26	415±27	1049±9	340	353	1054	336	356	1067	0.99	1.17±0.07	Present work
$_{70}Yb^{170}$	84.3	93±10	95±10	1020±20							1.03	1.24±0.15	Stepic et al
		86±6	85±6	983±11	77	77	994	75	75	993	0.989	1.13±0.08	Karlsson et al
		89±8	88±7	1000±30							1.007	1.16±0.09	Present work
		101±10	112±20	1110±20							1.02	1.25±0.15	Stepic et al
		90±4	96±4	1090±20							1.093	1.09±0.14	Graham et al
$_{74}W^{182}$	100.9	98±7	108±7	1080±20	82	89	1080	80	89	1093	0.994	1.21±0.08	Karlsson et al
		118±16	127±16	1072±21							0.988	1.43±0.18	Present work

volved, the data were analyzed by cutting off the tails at points such that the same percentage of area is contained in each peak. Thus part of the tail is omitted.

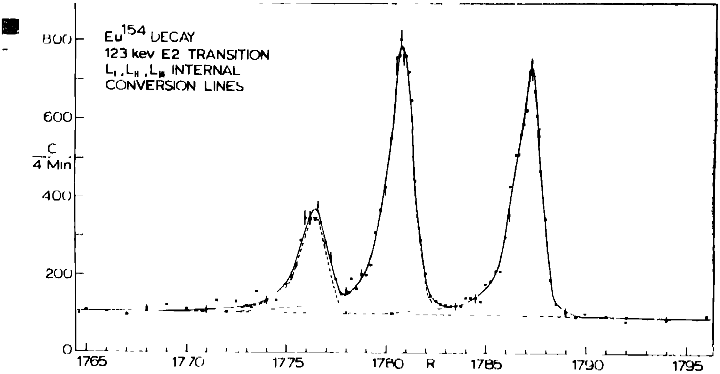

Fig. 4 A typical measurement of the L subshell lines of the 123 KeV transition in Gd^{154}

Various cutoff points below the peaks were used to check on the consistency of the analysis. Ratios obtained from these different cutoff points agreed with each other to within 3-4 percent One of the measurements is shown in figure 5. As in the case of Gd^{154} the L_3 line was used as the standard shape in order to separate

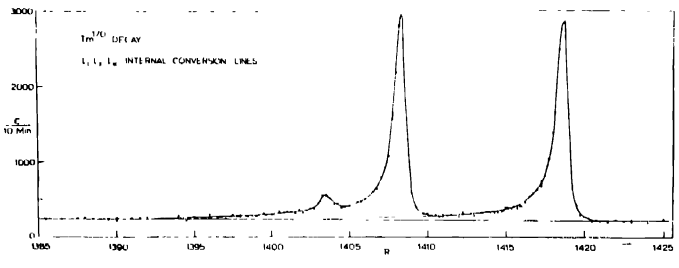

Fig 5. A typical measurement of the L subshell lines of the 84 KeV transition in Yb^{170}.

L_1 and L_2. The result also presented in Table I for the mean percentage deviation compared to theory is 16 ± 9 higher than theory.

The L subshell lines of the 100-keV transition in W^{182} were measured three times and the results averaged. The resolution varied from about 0 06 percent to 0.09 percent. Again line tailing was a problem and the same procedure of analysis was used here as was used in the case of Yb^{170}. In addition it should be emphasized that the relative errors in this procedure are such that they tend to cancel when one determines ratios. Figure 6 shows one of these measurements. The results were 43 ± 18 higher than theory for the mean percentage deviation.

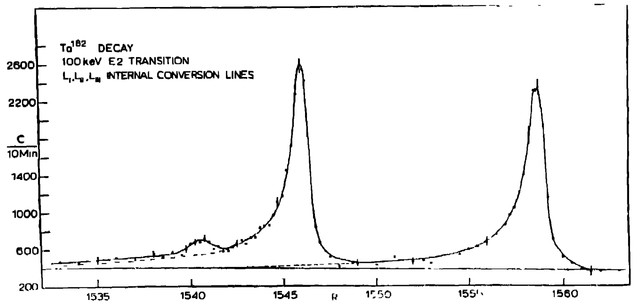

Fig. 6 A typical measurement of the L subshell lines of the 100 KeV transition in W^{182}

Figures 4, 5 and 6 do not show the complete energy range that was measured. Part of the background on either side of the peaks is omitted to better illustrate the peaks in the figures

The results are presented in Table I along with the theoretical values of Rose and Sliv and Band. In this Z region, Rose's and Sliv and Band's values agree closely with each other The results of other measurements in these isotopes are also presented in Table I. The L_1/L_2 ratios for Yb^{170} and W^{182} agree with the results of Stepic, Bogdanovic and Mladjenovic and Karlsson *et al* within the limits of experimental error. The results of Graham and Geiger are lower, however

In the time since the International Conference on the Internal Conversion Process at Vanderbilt University there has been more thought and study of the deviations from theory of the L-subshell ratios for E2 transitions in the rare-earth region. Figure 7 shows a plot of experimental measurements which are the sum of L_1/L_2 and L_1/L_3 from experiment divided by the average of this sum for the two theories, versus atomic number. This figure includes results presented at the Vanderbilt Conference by Stepic *et al*, Novakov *et al*. Graham and Geiger, Karlsson *et al*, and Brantley *et al*, and the published data of Herrlander and Graham (1964). Lines are shown in Figure 7 and represent $a \pm 6$ percent error in theory which includes the claimed $\lesssim 3\%$ uncertainty in the tabulated theoretical coefficients and $\lesssim 3\%$ uncertainty in the interpolation. All of the experimental ratios are higher than the corresponding theoretical values. This disagreement is small ($\lesssim 5\%$) in the heavy, spherical nuclei and within the errors of the theory. But the more deformed nuclei the discrepancies are, with one exception, outside the 6 percent error ascribed to theory and range up to 25 percent.

One notes, however; that the points with the largest discrepancies also have the largest uncertainties. The Chalk River group has considered this fact and

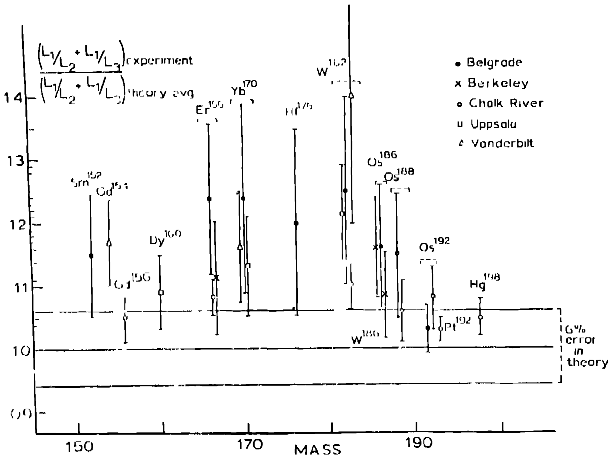

Fig 7. Experimental values of $L_1/L_2 + L_1/L_3$ divided by the averages of the theoretical values of Rose and of Sliv and Band. (The laboratories correspond to the following references; Belgrade, Stepic et al, 1965; Berkeley, Novakov and Hollander; Chalk River, Graham and Geiger and Herrlander and Graham, 1964; Uppsala, Karlsson et al, 1965; Vanderbilt, this paper)

feels that improvements in the accuracies of the measurements will bring theory and experiment into agreement within 6 percent. In order to clarify the differences between different experiments and between theory and experiment additional measurements and a careful study of the techniques of analyses should be made.

Further light is shed on this problem by studies of Hamilton *et al* (1965) on two deformed heavy elements Th^{228} and Pu^{240}. There the L_1/L_2 and L_1/L_3 results agree well with Sliv and Band but are 10-30% lower than those of Rose. This suggests that these effects may be related to the calculations of the weak L_1 conversion coefficient and not to some nuclear structure effect.

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